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Methods for measuring diffusion coefficients of radon in building materials

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Abstract

Two methods for determining the Rn-222 diffusion coefficient in concrete are presented. Experimentally, the flush and adsorption technique to measure radon release rates underlines both methods. Theoretically, the first method was developed for samples of cubical geometry. The radon diffusion equation was solved for boundary conditions imposing zero flux conditions successively on each side. In practice, a 100% effective covering would ensure this condition to be satisfied. The diffusion coefficient is then determined by comparing the computed and respectively measured normalized ratios of the radon release rates with respect to the rate corresponding to the open-boundaries (uncovered) specimen. As in practice, none of the investigated coverings showed to be effective in reducing radon exhalation, indicating that radon-tight sealing of surfaces is far from trivial, no clear conclusions could be drawn with respect to the diffusion coefficient. The second method can be applied to specimens that are first reshaped into hollow cylinders. A one-dimensional situation can be inforced by requiring that the flux vanishes at the two ends of the hollow cylinder. The theoretical and experimental ratios are again compared. In practice, the radon flux originates from a radon source enclosed inside the hollow cylinder, the effectiveness of the sealing being previously tested on an aluminum dummy of similar dimensions. The radon bulk diffusion coefficient for the used concrete sample resulted in a value of $D = (4.6 \pm 0.4) \times 10^{-6}$ cm^2 s^{-1}.

Keywords: Radon; Diffusion coefficient; Concrete; Release rate; Measurement method

1. Introduction

The diffusion coefficient of radon gas in building materials is often used as an indication for radon transportability through a porous medium and, furthermore, as an essential tool for quantitative predictions of radon concentrations in dwellings by estimating the exhalation rates of the wall surfaces. Between different building materials, there is a large range of values of this diffusion coefficient covering almost four orders of magnitude (Folkerts et al., 1984; Put and van der Graaf, 1996). Moreover, also within one type of material the variation in values is almost equally large. For example, for concrete, values are found (under laboratory or in situ conditions) in the range 7×10^{-10} to 1.5×10^{-7} m^2 s^{-1} (Folkerts et al., 1984; Renken and Rosenberg, 1995; Gadd

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and Borak, 1995). One has to be careful in comparing the values of the reported diffusion coefficients as the variations may occur due to a lack in uniformity with respect to their definition (Culot et al., 1976; Rogers and Nielsen, 1997), expressed either on the basis of bulk volume or pore surface area. However, the most likely cause is that of concrete being a complex and heterogeneous material, having its physical structure strongly dependent on the type of cement that is used, water cement ratios, curing conditions (Culot et al., 1976) and aging. For example, (Rogers et al., 1994, 1995) showed that, when corrected for density ifferences, the value of the diffusion coefficient for the aged concrete is approximately two times higher than the values for newly poured concrete.

From the existing methods to determine radon diffusion coefficients in building materials, the commonly employed one is by enclosing a slab of the material between two compartments: one kept at a high radon concentration while the other (initially) at low concentration. The radon diffusion coefficient is then deduced by either monitoring the radon in-growth in the (initially) low-concentration compartment or by measuring the steady-state radon flux into the compartment kept at a constant low (zero) concentration. In the first case a solution of the time-dependent radon transport equation (Zapalac, 1983) and in the second case, a steady-state solution (Folkerts et al., 1984) is used to calculate the diffusion coefficient. This method has the following drawbacks:

1. Radon-tight sealing between the slab and the compartment is very difficult to ensure and the presence of small leaks may lead to erroneous results.
2. Small pressure and/or temperature differences between the compartments may lead to additional transport due to advection and/or convection.
3. Sample dimensions are different from standard test samples in the building industry (test cubes, bars or cylinders).

In search of better methods that are based on a consistent terminology of the diffusion coefficient and circumvent some of the drawbacks of the existing techniques, two new methods for determining the radon diffusion coefficient of laboratory specimens of building materials are considered in this paper. We will elaborate on the theoretical basis underlying the methods, present results on concrete test samples for two geometries (cubical and cylindrical) and will discuss the domain of validity and the inconveniences of these methods. Both methods have the advantage that diffusion coefficients can be deduced from well-controlled radon release rate measurements of entire samples using a flush and adsorption technique, the measurement taking place after equilibrium is established (steady-state).

2. Theory

In a description of radon transport through a porous medium, information is needed on: radon generation, the structure of the medium, the physical processes causing the transport (driving forces) and the interaction of radon with the medium (partitioning between air and water phase and adsorption at the solid interface). This information may be conveniently contained in a single multi-phase time-dependent effective equation [Eq. (1)] for radon transport (Rogers and Nielsen, 1991; Van der Spoel, 1998), where the change in radon activity is expressed as the result of changes due to diffusion (first term right hand side), advection (second term), decay (third term) and generation (last term):

$$\beta \frac{\partial C_a}{\partial t} = \nabla \cdot (D \nabla C_a) + \frac{K}{\mu} \nabla P \cdot \nabla C_a - \beta \lambda C_a + S \tag{1}$$

with:

$\beta =$ partition-corrected porosity, $\beta = \varepsilon(1 - m + Lm) + \rho_b k_a$;
$\varepsilon =$ total porosity (air phase and water phase);
$m =$ fraction of water saturation of the pores;

L = Henry's law constant describing partitioning of radon between air and water phase, $C_w = LC_a$ ($L = 0.26$ at 293 K);
ρ_b = bulk density of dry material (kq m^{-3});
k_a = radon surface adsorption coefficient (m^3 kg^{-1}), $C_s = k_a C_a$;
C_a = radon concentration in air-filled pore space (Bq m^{-3});
C_w = radon concentration in water-filled pore space (Bq m^{-3});
C_s = adsorbed radon activity per unit of dry mass (Bq kg^{-1});
D = *bulk* diffusion coefficient (m^2 s^{-1});
K = intrinsic permeability of the material (m^2);
μ = dynamical viscosity of air ($\mu = 1.83 \times 10^{-5}$ Pa s);
P = pressure disturbance field relative to atmospheric pressure (Pa);
λ = decay constant of radon ($\lambda = 2.1 \times 10^{-6}$ s^{-1}); and
S = radon production per unit bulk volume (Bq m^{-3} s^{-1}).

Often used in the radon literature is the diffusion length ℓ (m), parameter related to the bulk diffusion coefficient according to,

$$\ell = \sqrt{\frac{D}{\beta\lambda}} = \sqrt{\frac{D_e}{\lambda}} \tag{2}$$

with D_e the so-called *effective* diffusion coefficient.

In this study we will make the simplifications that all situations considered are stationary, isothermal and isobaric, materials are homogeneous and that adsorption of radon to solid surfaces is negligible. Under these conditions Eq. (1) simplifies to,

$$D\nabla^2 C - \beta\lambda C + S = 0 \quad \text{with}$$

$$\beta = \varepsilon(1 - m + mL) \tag{3}$$

where the subscript a from the radon concentration in the air filled pore volume was dropped. This equation will be analytically solved for two geometries, namely a cubical and a cylindrical.

If the porous medium has a surface A through which radon is released, the bulk radon release rate, R(Bq s^{-1}), (total amount of released radon per unit time) is calculated by integrating the radon flux over the surface A according to:

$$R = -AD\nabla C. \tag{4}$$

where ∇C is calculated from Eq. 3

3. Cubical geometry

For a cubical geometry with L the length of the cube side (the method can also be applied for rectangular samples, differences in the theoretical approach will occur because of unequal sizes L_x, L_y and L_z), one starts by solving Eq. (3) for $C(x,y,z)$ with the boundary conditions:

$$C(x,y,z) = 0$$

$$\begin{cases} x = 0 & 0 \le y \le L, \quad 0 \le z \le L \\ x = L & 0 \le y \le L, \quad 0 \le z \le L \\ y = 0 & 0 \le x \le L, \quad 0 \le z \le L \\ y = L & 0 \le x \le L, \quad 0 \le z \le L \\ z = 0 & 0 \le x \le L, \quad 0 \le y \le L \\ z = L & 0 \le x \le L, \quad 0 \le y \le L \end{cases} \tag{5}$$

In principle, the solution depends not only on the diffusion coefficient D but also on the partition corrected porosity, β and the radon production rate, S. However, these last two parameters occur as a constant factor in front of some function f that depends only on D, the size of the cube, L, and the three coordinates: x, y and z,

$$C_0(x,y,z) = \frac{S}{\lambda\beta} f_0(D,L,x,y,z) \tag{6}$$

where subscript 0 denotes that no surfaces are covered. Furthermore, using Eq. (4), the corresponding radon release rate, R_0, (Bq s^{-1}) can be computed,

$$R_0 = 2D\left(\int_{y=0}^{L}\int_{z=0}^{L}\frac{\partial C_0}{\partial x}_{x=0}\mathrm{d}y\ \mathrm{d}z + \int_{x=0}^{L}\int_{z=0}^{L}\frac{\partial C_0}{\partial y}_{y=0}\mathrm{d}x\ \mathrm{d}z + \int_{x=0}^{L}\int_{y=0}^{L}\frac{\partial C_0}{\partial z}_{z=0}\mathrm{d}x\ \mathrm{d}y\right). \quad (7)$$

In the expression for R_0 the dependency on the three coordinates vanishes but the mathematical form is similar to the expression for C_0,

$$R_0 = \frac{S}{\lambda\beta}g_0(D,L) \quad (8)$$

This form led us to the idea to generate ratios, such that the constant factor in front would cancel. A possibility to achieve this is by successively covering faces of the cubical sample until it is totally covered. Mathematically, this means solving Eq. (3) but changing the boundary conditions (Eq. 5), i.e. imposing successively that the flux corresponding to the covered surface would vanish.

More precisely, let R_1 be the radon release rate obtained by covering the $x = 0$ face of the cube only, i.e. by changing the boundary condition at $x = 0$ into $\frac{\partial C(x,y,z)}{\partial x} = 0$, then $\frac{R_1}{R_0}$ would be such a ratio. Similarly, we can define R_2 (for the case when both the $x = 0$ and $x = L$ faces are covered), R_3 (also the face $y = 0$ gets covered), R_4 (flux on $y = L$ vanishes as well) and, similarly, for the z coordinate, obtaining R_5 and, R_6 respectively. Ratios are then calculated of all radon release rates with respect to the radon release rate corresponding to the uncovered cube. The logic of picking exactly this order of covering the faces is that the situation for the case when two opposite faces are covered is reduced to a two-dimensional problem while four sides covered corresponds to a one-dimensional case. A convenient way to obtain the solution in the n-dimensional case is by writing it as the sum of the solution of the $(n-1)$-dimensional case with uncovered boundaries and the solution in n-dimensions for Eq. (3) without the source term S with identical boundary conditions as the initial n-dimensional case (Carslaw and Jaeger, 1959).

The boundary conditions given by Eq. (5) may be applied to a sample in a laboratory in case this is enclosed in a measuring chamber permanently flushed with a radon free gas at such a rate that the radon concentration in the chamber is essentially equal to zero. The successive requirement of a zero flux on the faces of the cube implies that a 100% effective covering against radon should be used.

The calculated radon release ratios, $\frac{R_n}{R_0}$ (with $n \in [0,6]$) are plotted (Fig. 1) against the corresponding number of covered faces, n, for a cubical sample of side 0.15 cm (typical for standard test samples manufactured in the industry) and for (bulk) diffusion coefficients in the range 10^{-11} to 10^{-7} $\mathrm{m^2\ s^{-1}}$ which is typical for concrete (Renken and Rosenberg, 1995). The corresponding range for the diffusion length, ℓ, is 0.22–22 cm. For values at the lower end of this range (D is 10^{-11} $\mathrm{m^2\ s^{-1}}$; ℓ is 0.22 cm) radon release is a surface effect only, and scales with the number of covered surfaces. For values at the higher end of the range (D is 10^{-7} $\mathrm{m^2\ s^{-1}}$; ℓ is 22 cm), radon may exhale from the entire volume of the cube leading to the exhalation rate being independent of the number of covered surfaces until all surfaces are covered. At this point ($n = 6$), the radon release rate drops to zero. Both features are reflected in Fig. 1 by the linear and block shape of the curves for low and high values of the bulk diffusion coefficient, respectively.

4. Cylindrical geometry

In case samples of hollow cylindrical geometry with sealed top and bottom surfaces are used, Eq. (3) may be rewritten in cylindrical coordinates and for one-dimensional radial diffusion:

$$\frac{\partial^2 C}{\partial R^2} + \frac{1}{R}\frac{\partial C}{\partial R} - \alpha^2 C = 0 \quad (9)$$

with R the radial coordinate (m) and the newly introduced notation, $\alpha = \sqrt{\lambda\beta/D}$. A change in variable $r = R\alpha$ transforms Eq. (9) in a modified Bessel equation of order zero:

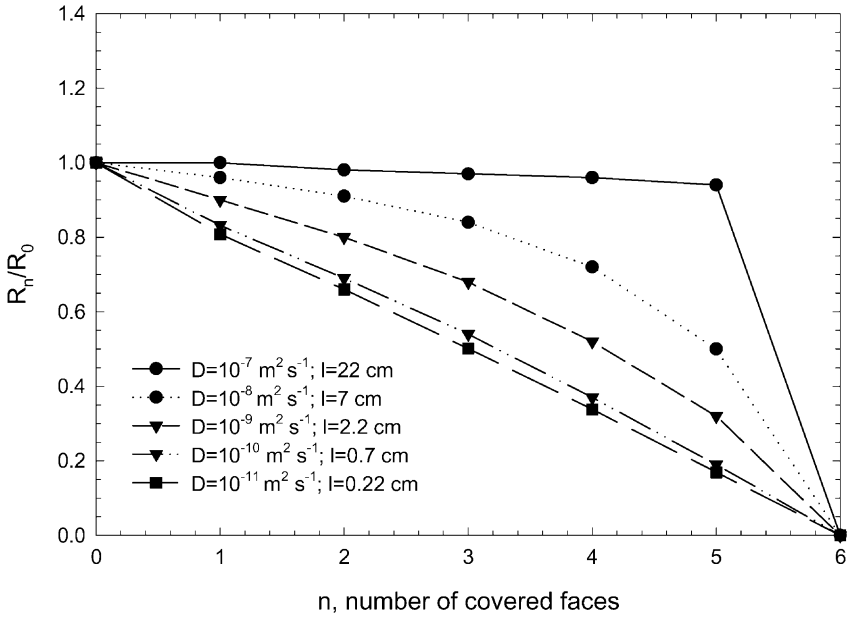

Fig. 1. Theoretical predictions of radon release rates ratios for the uncovered and covered conditions $\frac{R_n}{R_0}$ vs. number of covered faces (n) for different values of the bulk diffusion coefficient, D and the corresponding value of the diffusion length, ℓ assuming a 100% effective covering.

$$\frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r} - C = 0 \tag{10}$$

the solution of which is:

$$C(r) = AI_0(r) + BK_0(r) \tag{11}$$

where A and B are two arbitrary constants and I_0, K_0 the zero-order modified Bessel functions (Bowman, 1958). In practice a radon source with a strength R_{source} (Bq s^{-1}) that will generate a high diffusive flow of radon will be sealed in the hollow part of the cylinder. By knowing the volume of the sample, the values in Bq s^{-1} of the source strength and that of the radon production, S, can be compared ($S \ll R_{\text{source}}$) justifying the ommitance of S in Eq. (9). A solution of Eq. (10) is sought for that satisfies,

$$\begin{cases} r = a\alpha & C(r) = \dfrac{R_{\text{source}}}{\lambda V} + D\alpha \dfrac{\partial C}{\partial r_{r=a\alpha}} \dfrac{2\pi ah}{\lambda V} \\ r = b\alpha & C(r) = 0 \end{cases} \tag{12}$$

where a, b and h are the inner radius, outer radius and height of the hollow cylinder, respectively, and $V = \pi a^2 h$ is the volume of the inner cylinder.

The first boundary condition signifies that the value of the radon concentration inside the hollow cylinder is determined by generation from the source and loss via decay and diffusive flow through the walls of the cylinder. The second boundary condition implies that, at the external surface, the radon concentration is kept zero. The concentration $C(R)$ resulting for solving Eq. (10) is given by,

$$C(R) = \frac{R_{\text{source}}}{\lambda} \times \frac{I_0(b\alpha)K_0(R\alpha) - K_0(b\alpha)I_0(R\alpha)}{[K_0(a\alpha)I_0(b\alpha) - K_0(b\alpha)I_0(a\alpha)] + 2\beta/a\alpha[K_0(b\alpha)I_1(a\alpha) + K_1(a\alpha)I_0(b\alpha)]} \tag{13}$$

The radon release rate of the cylinder with the

enclosed source, $R_{\text{cyl}-\text{source}}$, [Eq. (4)] is found by multiplying the external lateral area of the hollow cylinder, $A_l = 2\pi bh$, with the bulk diffusive flux at $R = b$,

$$R_{\text{cyl-source}} = -A_l D \frac{\partial C}{\partial R_{R=b}} \tag{14}$$

Performing the differentiation in Eq. (14) finally leads to,

$$\frac{R_{\text{cyl-source}}}{R_{\text{source}}} = \frac{2b\beta}{\alpha a^2} \times \frac{I_0(b\alpha)K_1(b\alpha) + K_0(b\alpha)I_1(b\alpha)}{[K_0(a\alpha)I_0(b\alpha) - K_0(b\alpha) \times I_0(a\alpha)] + 2\beta/a\alpha[K_0(b\alpha) I_1(a\alpha) + K_1(a\alpha)I_0(b\alpha)]} \tag{15}$$

If the ratio on the left hand side of Eq. (15) is available from experiments, the equation can be solved for α (and thus D) provided that the partition corrected porosity is known, e.g. from an independent measurement.

5. Materials and methods

5.1. Samples

Investigations were conducted on concrete test cubes (15 cm side and an average mass of 8.5 kg) manufactured by ENCI (IJmuiden, The Netherlands) with a composition (standard procedure PBO-01) representative for the most widely used concrete in the Dutch building practice (Table 1). After being poured, the concrete cubes were stored (at ENCI) under water for 21 days and thereafter, for 7 days, in a laboratory facility under controlled temperature (20°C) and relative humidity (65%). Subsequently to their arrival at NGD-KVI, the cubes were wrapped in plastic foil and stored in a plastic bag for at least 1 month, to allow the curing process to continue. Periodical weighing of the test cubes shows a maximum loss of 0.6% in mass before the radon-release rate measurements started.

Table 1
Composition (per cube) of concrete mixture for the test samples

Component	Type	Mass (kg)	Fractional mass (%)
Cement	Blast furnish (IJmuiden)	1.22	14.4
Aggregates	Sand	2.41	28.4
	Gravel	4.28	50.5
Water	–	0.59	6.7
Total		8.5	–

For one of the cubes the radium content C^{Ra} and emanation factor η were estimated by sealing crushed parts of concrete in a 1-l Marinelli beaker and recording a γ-spectrum after allowing ^{226}Ra to reach secular equilibrium with the short-lived decay products (^{214}Pb and ^{214}Bi) of radon. A ^{226}Ra activity of 22.1 ± 0.6 Bq kg^{-1} was then deduced from decay of ^{214}Pb (295 and 392 keV γ-rays) and ^{214}Bi (609, 1120 and 1764 keV). By repeating the γ-measurement on the same Marinelli beaker without sealing, a radon emanation factor of $\eta = (0.18 \pm 0.03)$ was obtained.

For the second method, a hollow cylinder was carved out of one of the concrete cubes. After processing, the internal and external radius and the height of the hollow cylinder were measured (10 times) with a caliper: $a = (24.96 \pm 0.16)$ mm; $b = (67.1 \pm 0.3)$ mm and $h = (150.0 \pm 0.6)$ mm. The volume of the hollow cylinder resulting from these dimensions is $V_{\text{cyl}} = (1.83 \pm 0.02) \times 10^{-3}$ m^3.

To estimate the partition corrected porosity β, the total porosity, ε, and the fraction of water saturation of the pores, m, of the hollow cylinder were determined. The total porosity was calculated from measurements (after the radon release rate experiments were completed) of the wet mass m_{wet} (4.390 kg) and the dry mass, m_{dry} (4.168 kg), of the hollow cylinder using:

$$\varepsilon = \frac{m_{\text{wet}} - m_{\text{dry}}}{\rho_{\text{water}} V} \tag{16}$$

where ρ_{water} (kg m^{-3}) is the density of water. A

value of $\varepsilon = (0.1214 \pm 0.0011)$ was obtained. The mass of the hollow cylinder was also measured before the radon release rate determinations giving $m_{cyl} = 4.341$ kg. The fraction of water saturation m during the radon release tests on the hollow cylinder was estimated from:

$$m = \frac{m_{cyl} - m_{dry}}{m_{wet} - m_{dry}} \quad (17)$$

resulting in a value of $m = (0.779 \pm 0.006)$. Finally this leads to $\beta = (0.05142 \pm 0.0007)$ as estimate for the partition corrected porosity.

5.2. Radon source

A certified 20.2 kBq (4% uncertainty) radon-emanating source (Pylon, RN-2000A, serial no. 169) was used for calibration of the radon release rate setup and also in the second method for determination of the radon diffusion coefficient. The radon release rate of the source is $R_{source} = 0.0424 \pm 0.0017$ Bq s^{-1}.

5.3. Coverings

Epoxy glue (BISON Epoxy Rapide) — a universal two-component (a hardener and a raisin) quick hardening adhesive material — and *double-sided aluminized polyethylene foil* (TONZON thermosheet) — a 20-μm-thick membrane, developed and applied in the Netherlands for isolation purposes — were selected on the basis of a literature survey (Put and van der Graaf, 1996) and past experience at the NGD-KVI for covering experiments with the cubes. In addition, *soluble glass* (RUPLO HI, type 1530) — a silicon-based sealant, mixture of SiO_2 and NaO in water with volume proportions 2 ÷ 4:1 — was recommended as a material providing a very tight covering especially for concrete. The application and drying of the covers took place at room temperature, for all coverings no special pre-preparation of the concrete surfaces being performed.

Epoxies are known for having excellent adhesion properties, especially for fresh concrete (Ashton and Quirouette, 1984). Their reduction of radon-exhalation rates, when applied on concrete, varies between 30% and 93% (Van Dijk and de Jong, 1988; Quanlu and Hengde, 1998). The PE-foil was tested several times in our laboratory (Van der Spoel, 1998) indicating a bulk diffusion coefficient of $(1.1 \pm 0.4) \times 10^{-15}$ m^2 s^{-1}, sufficiently low to qualify the membrane as an almost 100% effective radon barrier. Silicone coatings are indicated as a good radon barrier (Put and van der Graaf, 1996), silicon-based caulks are found to attenuate ^{222}Rn by 75–98% (Fleischer, 1992).

5.4. Radon release rate measurements

Radon release rates of test samples were measured, under controlled conditions, with the NGD-KVI radon release rate setup (Fig. 2) consisting of a stainless steel box (80 l) inside which a test sample (BM) is placed. A nitrogen flow of

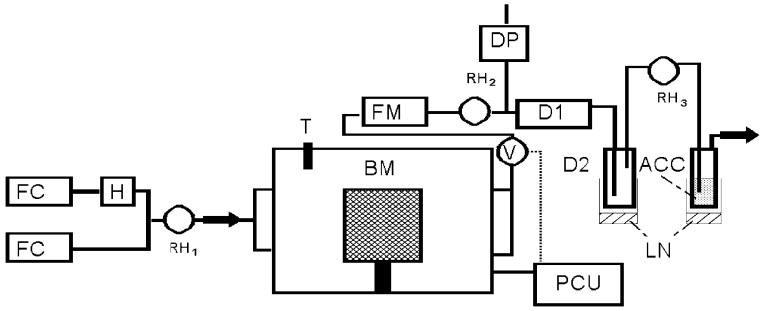

Fig. 2. The NGD-KVI exhalation setup. FC, flow controllers; H, humidifier; RH_1, RH_2, RH_3, relative humidity sensors; T, temperature sensor; BM, building material; V, control valve; PCU, pressure control unit; FM, flow meter; DP, differential pressure transducer; D1, silica gel (pre-dryer); D2, water trap; LN, liquid nitrogen; ACC, 17.1 g of activated charcoal at LN temperature during the measurement time.

400 ml min^{-1} conditioned to 50% relative humidity passes through this box. The pressure inside the setup is kept constant to a preset value (1060 Pa) by a feedback system (PCU) to prevent fluctuations due to atmospheric pressure variations. To achieve steady-state conditions inside the exhalation volume, the sample is pre-flushed for at least 17 h before the measurements take place. Supplementary, the temperature variations are also monitored (T). The value of the outgoing flow together with its relative humidity are measured (FM and RH2, respectively) yielding information about a possible change in humidity of the sample. This flow is being pre-dried with a column of silicagel (D_1) and freeze-dried via a vial cooled to liquid nitrogen temperature (D2, LN) thereafter passing through a column of activated charcoal (ACC) that adsorbs the radon exhaled by the sample and transported with the nitrogen flow.

After adsorption, the charcoal is transferred into a cylindrical pill box and placed in a calibrated geometry on one of the NGD-KVI low-background high purity germanium spectrometers (EG&G ORTEC) for a γ-ray measurement. The count rate is determined in the energy range of 50 keV to the upper energy limit of the spectrometer. The adsorbed radon originating from the test sample N_{bm} is related to the total count rate, N_{tot} (s^{-1}), in the energy range of the spectrometer. Corrections for the contribution of the charcoal, N_{bg} (s^{-1}), for radon decay during the measurement time of the gamma-ray determination, t_m (s) and for the time interval [waiting time, t_w (s)] between the end of the adsorption and the beginning of the gamma measurement, are applied. The contribution of the charcoal, was computed from the recorded spectrum of a standard amount of activated charcoal under the same geometry. The waiting time, t_w (s), was always longer than 3 h to allow radon to reach secular equilibrium with its gamma-emitting descendents. The exhalation rate, R (Bq s^{-1}), of the building material is then computed (Van der Graaf et al., 1998) from:

$$R = \frac{N_{bm}\lambda(\lambda_v + \lambda)}{\omega\lambda_v(1 - e^{-\lambda t_a^{bm}})} - R_{setup} \quad \text{with}$$
$$N_{bm} = (N_{tot} - N_{bg})\frac{\lambda t_m e^{\lambda t_w}}{1 - e^{-\lambda t_m}} \tag{18}$$

where:

λ, λ_v = decay constant of radon and, respectively, ventilation rate of the setup (defined as the ratio between the flow and the volume of the box) (s^{-1});

ω = efficiency of the method, a product between the fraction of radon adsorbed on the charcoal and the efficiency of the gamma detection system; and

R_{setup} = contribution (Bq s^{-1}) of silicagel in the pre-dryer of the setup. This contribution $(2.9 \pm 0.4) \times 10^{-6}$ Bq s^{-1}, is small compared to release rates of concrete.

The efficiencies of the spectrometers were determined by conducting release rate experiments with the certified radon-emanating source. Typically these efficiencies are in the range 0.16–0.26.

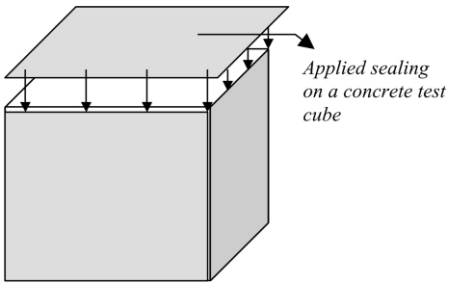

Fig. 3. Application of face-by-face sealing on concrete test cube.

5.5. Measurement method for samples of a cubical geometry

Samples of cubical geometry are enclosed in the volume of the setup for radon release rate determinations. Next, a successive covering of the faces of the cube with one of the previously mentioned coverings is performed (Fig. 3). After each step, the radon release rate is measured at

least three times and the ratios $\frac{R_n}{R_0}$ (with $n \in [0,6]$) are computed. By comparing the experimentally obtained plot of ratio against number of faces covered with the theoretical one, the bulk diffusion coefficient can, in principle, then be determined.

5.6. Measurement method for samples of a cylindrical geometry

After reshaping the sample in a hollow cylinder, its radon release rate is measured. Next, a radon source is enclosed inside the hollow cylinder (Fig. 4) and the measurement is repeated. The effectiveness of the sealing of the radon source inside the hollow cylinder — important to ensure the condition that the radon flux at the top and bottom surface is zero — was tested, by using the same procedure, on an aluminum dummy. To allow the concentration inside the hollow cylinder to reach steady-state conditions, a time interval of 3 weeks has to pass before the radon release rate determination. Using the experimental values of the ratio $\frac{R_{\text{cyl-source}}}{R_{\text{source}}}$ and of β in Eq. (15) the bulk diffusion coefficient can be determined.

6. Results and discussion

6.1. Cubical geometry

Fig. 5 shows the results of the radon release rate measurements for a concrete cube whose faces are successively covered with double-sided aluminum polyethylene foil attached to the surfaces using epoxy glue. According to the theoretical predictions (assuming a 100% effective covering), the ratios $\frac{R_n}{R_0}$ (with $n \in [0,6]$) should decrease as a function of the number of faces covered. However, the experimental ratios are approximately independent of n and their value does not significantly differ from unity. The most surprising fact is that even the measurement on the completely covered cube does not result in any decrease in the release rate. Comparison of the theoretical and the experimental curves shows that either the assumption of the combined covering (epoxy and PE-foil) being 100% effective is not satisfied or that small leaks have been present at the edges after application of the foil. The latest, in combination with a large bulk diffusion coefficient (implying a diffusion length of radon in concrete larger than the average distance from the point radon was produced to such openings), would deteriorate the covering effect. Results obtained for a second test cube (Table 2) completely covered with one continuous layer of epoxy glue followed by a subsequent application of a complete cover of the aluminized PE-foil on the top of the first layer, show that the epoxy seems to form a layer that acts as a path allowing for radon to diffuse easily (the foil is supposed to be radon tight). This hypothesis was once again supported by an additional experiment in which a radon source was inserted between two sheets of foil that where joined at their edges with epoxy glue. In a radon release measurement on this 'wrapped' source no significant reduction of the source strength was found.

To further avoid this situation, i.e. to prevent radon escaping through an intermediate layer

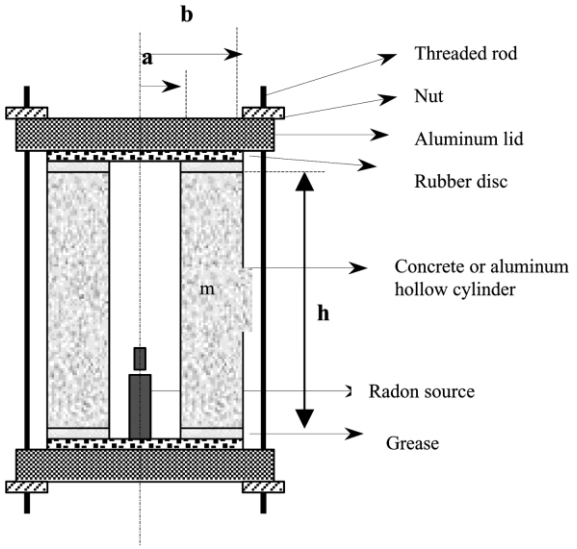

Fig. 4. Concrete or aluminum hollow cylinder with a radon source enclosed.

Table 2
Radon release rates, R, of a concrete cube totally covered with one layer epoxy glue and with an additional layer used as adhesive for the double-sided aluminized polyethylene foil[a]

Sample	R (Bq s^{-1})	Reduction (%)
Bare cube	$(4.63 \pm 0.15) \times 10^{-5}$	–
One layer epoxy glue	$(3.66 \pm 0.07) \times 10^{-5}$	14
Additional aluminized PE-foil	$(2.41 \pm 0.05) \times 10^{-5}$	48

[a]The reduction with respect to the release rate of the uncovered cube is also indicated.

between the cover and the concrete, a one-component cover, soluble glass (known to penetrate the superficial layer of the sample surface and form an elastic film after hardening such as no cracks or small openings are expected to appear during drying) was next used. Measurements for another bare, one layer and three layers of soluble glass covered concrete cube are summarized in Table 3. The superposition of three layers reduces the radon release rates with only 24%. Moreover, from a visual inspection it appeared that if in the beginning the soluble glass forms a glossy surface, a severe degradation within a time span of several months occurs. It is speculated that the reaction of soluble glass with the atmospheric CO_2 creates an acid environment, which allows the soluble glass to react afterwards with the silicate and carbonate groups present in concrete. In the first stage of the reaction, a gel that consists of water and a large number of agglomerates connected with the Si–O–Si bridges is formed. In time, the gel dries and only a strongly shrunken layer of sand (SiO_2) remains. The corroded surface looses all its properties as a radon barrier.

Clearly, with the coverings investigated up to now no conclusion was reached with regard to the radon bulk diffusion coefficient. Our experience shows that covering of concrete to inhibit radon release is not a trivial thing. When trying to apply this method attention should be taken to the selection of coatings, application method, and possible interactions with the substrate and durability properties.

6.2. Cylindrical geometry

The results of the measurements of radon release rates on the aluminum hollow cylinder for

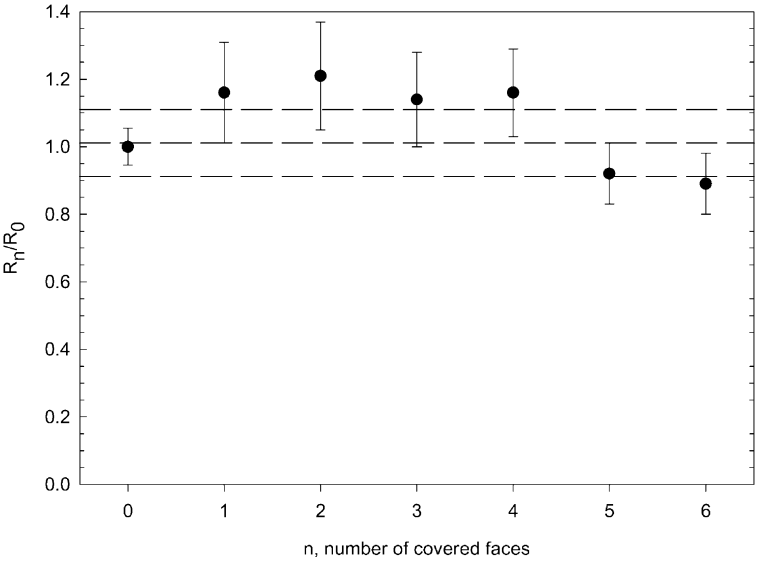

Fig. 5. Experimental dependence of ratios of radon release rates vs. the number of covered faces. A face-by-face covering of the surface of a concrete cube with a combination of epoxy glue and double-sided aluminized polyethylene foil was performed. The solid line represents the weighted mean ($\chi^2 = 1.3$). The dashed lines represent the weighted mean $\pm$ 1 S.D. (external error).

Table 3
Radon release rates, R, for a complete covering of a concrete test cube with one and three layers, respectively, of soluble glass

Sample	R (Bq s^{-1})	Reduction (%)
Bare cube	$(4.71 \pm 0.08 \times 10^{-5}$	–
One layer soluble glass	$(3.84 \pm 0.07 \times 10^{-5}$	19
Three layers soluble glass	$(3.59 \pm 0.06 \times 10^{-5}$	24

testing the effectiveness of the top and bottom sealing of the radon source (Table 4) show that the sealing is almost completely radon tight (94% effective). Furthermore, the radon release rate of the concrete hollow cylinder (Table 4) is found to be three orders of magnitude smaller than the release rate of the hollow cylinder with the enclosed radon source, a fact that justifies the assumption (see theory section) that the radon production can be neglected during the measurements. Assuming that the same effectiveness of sealing was achieved for the radon release rate measurement of the hollow cylinder with the radon source enclosed as for the aluminum dummy, the value for the ratio $\frac{R_{\text{cyl-source}}}{R_{\text{source}}}$ was calculated as: (0.374 ± 0.019). Using this ratio (Fig. 6) together with the value for the partition corrected porosity in Eq. (15), a value for the bulk diffusion coefficient is calculated as $D = (4.58 \pm$

Table 4
Radon release rates, R, for the aluminum dummy with enclosed radon source and for the concrete hollow cylinder with and without enclosed radon source

Sample	R (Bq s^{-1})	Observations
Aluminum dummy with radon source	$(6.04 \pm 0.22 \times 10^{-4}$	A 99% reduction sealing is obtained
Concrete hollow cylinder	$(2.00 \pm 0.05 \times 10^{-5}$	Radon production of hollow cylinder is much smaller that of the source
Hollow cylinder with radon source	$(1.58 \pm 0.15 \times 10^{-2}$	$R_{\text{cyl-source}}$

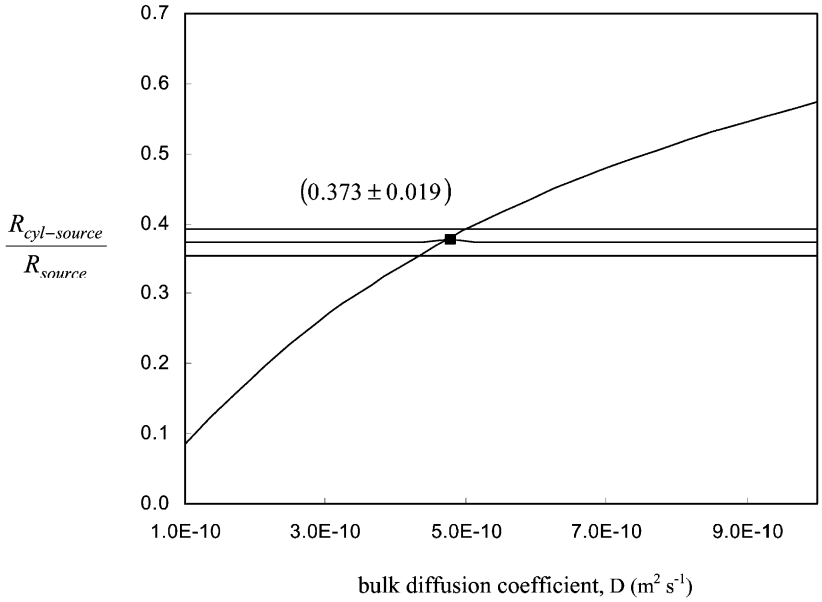

Fig. 6. Hollow cylinder experiment: ratio of radon release rates vs. bulk diffusion coefficient. The coordinate on the x axis corresponding to the intersection point for a certain experimental determined value of the ratio $\frac{R_{\text{cyl-source}}}{R_{\text{source}}}$ represents the value for the bulk diffusion coefficient of radon.

$0.35) \times 10^{-10}$ m^2 s^{-1}. An assessment of the propagation of uncertainties showed that the uncertainty in the experimental ratio is the main source of uncertainty in the diffusion coefficient. The result is less sensitive to the uncertainty in the partition corrected porosity.

The value of the diffusion length corresponding to the determined bulk diffusion coefficient is: $\ell = (6.5 \pm 0.3)$ cm. With respect to our previous analysis (Fig. 1), it can be concluded that radon release for this concrete results for a major part from release from the surface layer up to a few centimeters deep. Roughly, less than half of the radon that is generated at depths of 6.5 cm and deeper reaches the concrete surface and contributes to the radon release rate. Intuitively and also more empirically, this was also concluded after comparing the values of radon release rates of the concrete cube used for making the hollow cylinder, the hollow cylinder itself and crushed parts that remained after processing (Table 5). The fact that within the uncertainties, the radon release rates of the concrete cube and of the hollow cylinder are comparable, allows the assumption that during processing, no big alteration of the physical structure of concrete occurred (thus also no alteration of the diffusion coefficient). On the other hand, it appears that the radon release rate for the crushed parts (diameters smaller than approx. 1–2 cm) is more than a factor of 2 higher than for cube/hollow cylinder also indicating that in samples of this type of concrete with effective dimensions of 7.5 cm (cube) and even as small as 2.5 cm (hollow cylinder) a major (more than 50%) part of the generated radon is not released.

In principle, both methods should work for any type of building material, as far as a specimen of it can be shaped in cubes or hollow cylinders. The method developed for the case of a cubical geometry is limited by the fact that the values of the bulk diffusion coefficient D depend on the size of the test specimen. As also depicted in Fig. 1, for large values of D, the ratios of radon release rates $\frac{R_n}{R_0}$ or $\frac{R_{\text{cyl-source}}}{R_{\text{source}}}$ will not significantly differ from unity and thus only allow for lower limit determination of the value of the diffusion coefficient. This limitation can be circumvented by using larger specimens.

Due to the measurement principle underlying both methods, the drawbacks mentioned in Section 1 are surpassed. Other difficulties, however, occur in the way that the samples are shaped and prepared for the radon release measurement. The first method seems to be a general and straightforward way to determine the radon diffusion coefficient, the problem in finding a radon-tight covering makes it, up to now, impractical. The second method offers a satisfactory accurate determination of the diffusion coefficient but with the compromise of more assumptions and a peculiar geometry.

7. Conclusions and outlook

Two methods for measuring diffusion coefficients of radon in building materials were presented. The geometries of the investigated samples — standard Dutch concrete — were cubical, respectively hollow cylindrical. For these two geometries the radon transport equation was solved and the theoretical basis for calculation of the bulk diffusion coefficient from successive radon release rate measurements on the samples

Table 5
Radon release rates for original concrete cube, the hollow cylinder made from it and crushed concrete parts that remained after processing of the hollow cylinder

Sample	R (Bq g^{-1} s^{-1})	Observations
Concrete cube	$(4.33 \pm 0.19 \times 10^{-9}$	$m_{\text{cube}} = 8077$ g
Hollow cylinder	$(4.61 \pm 0.05 \times 10^{-9}$	$m_{\text{cyl}} = 4341$ g
Crushed concrete parts	$(1.01 \pm 0.13 \times 10^{-8}$	$m_{\text{crush}} = 1583.5$ g

was outlined. The radon release rates measurements are conducted under well-controlled steady-state conditions, using a flush and adsorption technique.

The method developed for a cubical geometry, although widely applicable and straightforward, fails because of the difficulties encountered in finding a 100% effective covering against radon. Research into finding such covers is presently still ongoing.

For the second method, where samples have to be reshaped as hollow cylinders, a satisfactory accurate determination of the diffusion coefficient was achieved for commonly used Dutch concrete. This method, however, requires more assumptions and a more complex geometry. Up to now this method has only been tested on one specimen and further validation on more samples is foreseen.

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